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Recently de Boissieu *et al.* proposed an explanation for the broadening of acoustic modes observed in quasicrystals (QC). It is the transcription of a well-known model used for glasses. We raise two fundamental objections against applying it to QC.

Recently, de Boissieu et al. [1] proposed a mechanism to explain the broadening of phonons starting from a threshold wavevector in QC, as is systematically observed in experimental data. The mechanism (the coupling of sound waves to a heath wave) is quite general and not new, as it has been known for many years in glasses. There is obviously no doubt that the mechanism itself is sound but we must take issue with the way it is being used through a transcription to the field of quasicrystals. Below, we explain our objections on two levels:

(1) On a general level, phonon broadening is an intrinsic property of quasicrystals, even in a completely harmonic model, such that there is *a priori* no need for the introduction of an assumption of anharmonicity in the form of a coupling.

(2) On a more detailed level, the authors try to blow new life into a cluster scenario proposed by Janot et al.[2], by using the idea of localized modes on clusters as a basic ingredient for the microscropic realisation of the mechanism. The localized modes on the clusters are this time no longer directly responsible for the broadening, but they are proposed to be flat modes, that couple to the sound waves. To cite them verbatim: "The building bricks of all QC structures are atomic clusters. These clusters are not mere geometrical constructions but real physical entities responsible for specific features in the QC vibrational spectrum (e.g., responsible for localized modes)".[3]

Hence on two levels assumptions are introduced that are not granted or even not needed, while through the presentation the reader might be left with the impression that the experimental data present evidence in support of these assumptions.

(1) There exists an extensive literature on phonons in QC, e.g. on the Fibonacci chain, based on the transfer matrix method.[4] What transpires from such (rigorous) studies is that the phonon eigenmodes are not at all periodic and even not quasiperiodic. (There exist e.g. so-called recurrent eigenmodes: As a function of its position in space the amplitude of a mode can exhibit humps around the values $a \times \tau^n$, where τ is the golden mean, n is the set of integers, and a a constant (length). In between the humps, the amplitude of the mode is never zero but it can be very weak). The simple argument of the non-quasiperiodicity of the eigenmodes shows that their Fourier analysis in terms of wave vectors Q will not show dispersion curves of zero line-width as in crystals, but broader features, and that this broadening is not in energy (as de Boissieu et al. think) but in the wave-vector Q. For the recurrent modes, specialists even wonder if it is a mathematically legitimate to take it for granted that they have a Fourier transform in Q-space, and in the case it is not, what kind of information the neutron data contain about these modes.

These features occur even in models that are perfectly harmonic without any coupling between the modes. Of course calculations in the harmonic approximation on successive approximants can never reveal such broadening, because they give rise to zero linewidths by definition. A key intrinsic difficulty of the problem is thus just being missed by such an approach, but it is from the blind spot inherent to such calculations that one might feel the urge to inject additional assumptions of anharmonicity into the problem under the form of localization or couplings.

It would be cheap to push the present objections away, by arguing that real QCs are not one-dimensional, and that surely there will be a loophole of escape from these objections, when we go to higher dimensions. Such vague arguments would (a) reverse the charge of proof, and (b) contain a tacit denial of the horrendous difficulty of eigenvalue problems (with the correct boundary conditions) on quasiperiodic structures. We may add to this that (c) anharmonicity can be experimentally evidenced by the temperature dependence of the Debye-Waller factor.[5]

When one drops full mathematical rigor, approximate eigenmodes that are periodic or quasiperiodic, leading to zero approximate line widths, are only expected to exist in the long-wavelength limit. This correlation between absence of broadening and long wavelengths is confirmed by the experimental observations. But the cluster mechanism proposed by de Boissieu et al. only holds in the limit in which acoustic wavelengths are much larger than a typical cluster size, i.e. the very limit where on the basis of the preceding arguments the broadening of the line widths is rather expected (and observed) to be minor.

(2) In key positions of their paper (the abstract and the final conclusion) the authors stress the important rôle they propose clusters to play in the microscopic realisation of their mechanism. The authors discuss the

relevance of *isolated* clusters, and present the issue in terms of a unique isolated spherical inclusion in a vibrating medium (e.g. a metallic sphere inside a rubber medium). They dismiss such a scenario with great emphasis, but this does not really clarify the assumptions that underly their paper with respect to the issue if there are isolated clusters in quasicrystals: The scenario chosen to assess this issue is too obviously wrong, and the real issue if there are isolated clusters in quasicrystals is more subtle.

We have to address here a difficult situation of possibly ambiguous terminology, because the authors do indeed introduce a concept of "isolated" clusters, different from the one that might be inferred from their presentation. Let us call the type of isolation evoked by the model of a *unique* spherical inclusion *"type 1"* and the type of isolated clusters used by the authors *"type 2"*. The inadequacy of the type-2 isolated cluster model does not hinge on the abundance of clusters in the structure as one might infer from the type-1 model with its unique cluster. It rather consists in tacitly denying the importance of boundary conditions in the set of coupled differential equations that describe the phonon problem (and which are in general expressed in terms of a dynamical matrix).

The point is easily understood as follows. Take e.g. a one-dimensional crystal that is based on the periodic repetition of the small motif $LSLLS$ taken from the Fibonacci chain. The crystal is thus $...LSLLS.LSLLS.LSLLS....$ Based on visual clues we could claim that $LSLLS$ is a cluster, and that the crystal is a dense packing of clusters. The eigenmodes and eigenvalues of an isolated cluster $LSLLS$ are completely different from those of the crystal $...LSLLS.LSLLS.LSLLS....$ In fact, the cluster $LSLLS$ has six discrete flat modes with a certain dynamical form factor, $S(\mathbf{Q})$ that extends throughout reciprocal space. The modes are thus flat due to the finite extension of the cluster in space. The modes of the crystal are completely different: They do not correspond to a few isolated discrete energies, but build a whole dispersion curve and for each eigenmode (i.e. each energy) in the acoustic regime the Q-dependence is Dirac-like in reciprocal space (if we limit ourselves to one Brillouin zone).

Of course, it would be completely inappropriate to claim on the basis of the visual clue that we can discern clusters $LSLLS$ in the crystalline structure that there are flat modes in the crystal, and that there would exist a coupling between sound waves and these flat modes in the crystal. The flaw in such a reasoning is uniquely based on a tacit change of the boundary conditions: It replaces periodic boundary conditions (with a rather smooth variation of the force constants across the "cluster" boundaries) by an abrupt discontinuity at the "surface" of the imaginary cluster.

In other words: Type-1 isolation refers to the absence of similar clusters in the surroundings, the cluster is alone. Type-2 isolation refers to a decoupling of the cluster from the surroundings in terms of the force constants. And of course what is relevant for the phonon problem is not if the cluster is alone (type-1 isolation) but how the clusters couple to their surroundings at their supposed boundaries (type-2 isolation). Even in the example of a unique metallic sphere in a rubber medium, it is type-2 isolation that is physically relevant.

QCs are not periodic, and are subject to other boundary conditions than the ones that prevail in a crystal (see below). But this certainly cannot mean that our example would not be appropriate and that the introduction of "clusters" by the authors would have less of a hidden problem with the prevailing boundary conditions, because the crucial point of our objection lies in the postulated abrupt discontinuity, not in the periodicity. When I am raising an objection against the use of (*type 2*) isolated clusters, it can thus certainly not imply that I would have missed the passage in the paper where the authors acknowledge that there are no (*type 1*) isolated clusters in quasicrystals. The statement that there are no (*type 1*) isolated clusters in QCs (because they are a dense packing of clusters) cannot hide the fact that it is the very use of (*type 2*) isolated clusters which is the basic ingredient for the microscropic interpretation proposed by the authors.

We could never warn the reader enough against the pitfall that would consist in getting one's attention sidetracked towards the issue if there is convincing evidence for the presence of clusters in QCs or otherwise. That would be certainly an interesting topic in its own right, but rather pointless and misleading in the present discussion, as the issue if there are (type-2) *isolated* clusters in quasicrystals cannot be replaced by an issue if there are clusters in quasicrystals all together, nor by the issue if clusters are physically meaningful in quasicrystals. The verdict on the latter issues will moreover depend on the context of the application: A possible cluster argument in a problem of stability or electronic properties will be different from the one in a phonon problem.

The authors formulate the statement that clusters are not mere geometrical constructions without any proof as though it would be an obvious thruth, and the difficulty that they can overlap is passed under silence. The claim that the origion of the flat modes observed in AlPdMn can be attributed to a localization on clusters is also put forward without any proof.

To introduce the boundary conditions underlying their cluster assumptions, the authors should have given arguments that there is a discontinuity in the force constants at the surface of these clusters. In certain points on the cluster boundaries, the contrary rather seems to be true, viz. when instead of being isolated clusters overlap, which is often the case. In such points it rather looks as though nothing in the whole set of the atomic forces between pairs of atoms in a QC singles out a cluster as an isolated entity, defined by such a discontinuity. The forces between the atoms inside the clusters are not obviously different from those between an atom of the cluster

and a neighbouring atom that lies just outside the cluster (but inside the overlapping cluster of the same type). A few phason jumps can create the illusion that a whole cluster has jumped, which also clearly illustrates the relative arbitrariness of assigning an atom to a cluster and of suggesting that a cluster would be an isolated entity whose existence would be obviously defined by a discontinuity in the atomic forces at its surface.

Mathematically spoken, if a cluster is taken large enough it can even be a covering cluster for the whole QC. One can imagine a crystal that could be depictured as a (periodic) arrangement of physically acceptable, overlapping identical clusters of a certain size, and that would not lead to any localization or broadening. Any attempt to escape from this trivial objection must therefore forcedly end up in a discussion of the global, non-periodic arrangement of the clusters and their overlaps.

Discussing QC problems in terms of clusters rather than atoms, is thus just a kind of renormalization procedure, that merely shifts the intrinsic difficulty of non-periodicity to a different length scale, but does not tackle the difficulty itself. It is a blunt denial of the subtlety and difficulty of the eigenvalue problem to overemphasize the rôle of clusters. We can illustrate this with the Fibonacci chain. It starts with *LSLLS.LSL.LSLLS.LSLLS.LSL.LSLLS.LSLLS.LLS....*, where we have subdivided the sequence in building bricks $LSLLS$ and LSL. Each of the ocurrences $.LSL.$ herein is seen to be followed by LS as both building bricks $.LSL.$ and $.LSLLS.$ begin with LS. Hence the whole sequence can be seen as made from the "covering cluster" $LSLLS$, whereby we have to allow for overlaps LS, which appear exactly at the positions where we have separated out $.LSL.$. Similarly we could even consider LSL as a covering cluster (the overlap would then be L). Now, the phonon eigenmodes of the isolated sequences LSL and $LSLLS$ can be calculated (from the corresponding 4×4 and 6×6 dynamical matrices). What does this handful of eigenmodes tell us about the phonons of the Fibonacci chain? Hardly anything! As we pointed out above, even the phonons of the periodic sequences based on LSL or $LSLLS$ do not give us the correct picture, despite the fact that in such sequences the clusters are no longer completely isolated (which would be a completely irrealistic boundary condition) and one at least allows for the point that they are *embedded* in a larger structure (which completely changes the eigenvalue problem).

The idea of clusters LSL and $LSLLS$ certainly has great eye appeal. One might think at first sight that it must yield great insight in the dynamics of the Fibonacci chain. But as we explained above, all this is mere deception. Already the overlap $LSLLSLLS$ of two clusters of the type $LSLLS$ will yield competely different solutions for the eigenvalue problem than $LSLLS$ itself. The same basic objections about the boundary conditions remain perfectly valid in the three-dimensional case, such that the fact that we work on the one-dimensional case does not present a loophole from these objections. All the use of the clusters LSL and $LSLLS$ allows us to do is to rewrite the transfer matrix formalism in terms of matrices that correspond to LSL and $LSLLS$ rather than in terms of the more elementary matrices that correspond to S and L. This illustrates how replacing atoms by clusters is just a renormalization procedure, as we stated. It is an underestimation of the complexity of eigenvalue problems and their boundary conditions (which is *global*) to suggest that they could be approached *locally* by focusing one's attention to small building bricks. Putting the bricks together just changes everything.

At least in the present context we can thus state that unless a rigorous proof of the contrary is given, it is wise to adopt cautiously the conservative view point that the rigorous application of the idea of clusters, even if they look physically attractive, has remained limited to just a convenient pictorial shorthand to describe parts of the structure, nothing more. We can appreciate from this discussion how both objections (1) and (2) are linked, in the sense that both are based on a tacit modification of highly sensitive details of an eigenvalue problem, that is very hard to spot. The example of how the recurrent modes completely escape the analysis in terms of periodic approximants, shows to what kinds of catastrophies such lack of rigor can lead. Once again, this concern about rigor should not be misrepresented by saying that I would claim that there are no clusters in quasicrystals, or that clusters could not play a role in quasicrystals, etc...

Without any justification, the localized modes invoked are identified with the flat modes that have been reported in AlPdMn, and a coupling mechanism between these localized modes and sound waves is proposed. We have two objections to this:

(a) Such an explanation for the flat modes is just one between several other possibilities. One of the alternatives is documented and can therefore not be ignored: By a scrutiny of the displacement patterns in their numerical simulations Hafner and Krajci [6] were able to associate the flat modes with a restriction ("confinement") of the vibrations to disclination lines of atoms that are topologically different from average (e.g. the atoms have a 13-fold coordination, rather than a normal 12-fold one). This has nothing to do with the vibration on a cluster.

(b) The issue if the flat modes are due to a localization on clusters is not open-ended within the present state of knowledge. It can be unambiguously settled. It suffices to check if the structure factor of a flat mode is indeed compatible with the dynamical structure factor of a cluster vibration (as Buchenau has done to prove his model for the dynamics of silica). Although the dynamical structure factors of the flat modes have not been published, it must be straightforward to extract this first-rank information from the authors' already existing data, and a numerical calculation of the vibrational spectrum of a Bergman or a Mackay cluster with realistic force constants, involving typically 33 to 55 atoms, is certainly

not unfeasible.

Hence, before one can formulate any possible approach of the type proposed by the authors, it is a peremptory prerequiste that one first proves on the basis of existing data, that (1) the observed structure factors of the flat modes are compatible with an interpretation of these modes in terms of cluster phonons, and (2) that there are anharmonicities within the system, e.g. on the basis of Debye-Waller factor anomalies of which one has proved beyond any doubt that they cannot possibly be attributed to an onset phason hopping. These are necessary but not sufficient, minimal conditions that have to be met. They stand completely free from any theoretical considerations, and therefore add up completely independently to the two main objections outlined in the present Comment.

[1] M. de Boissieu, R. Currat, S. Franconal and E. Kats, Phys. Rev. B **69**, 054205 (2004).

[2] C. Janot, A. Margl, B. Frick and M. de Boissieu, Phys. Rev. Lett. **71**, 871 (1993); C. Janot and M. de Boissieu, Physica B **219-220**, 328 (1996).

[3] (a) Our verbatim citation can be found on page 5, at the top of the second column of reference [1]. (b) The authors qualify the optic modes observed in QCs as "dispersionless". As acoustic phonons in the long-wavelength limit are also dispersionless, we prefer the terminology "flat", which not only indicates that the slope is constant with the wave vector **Q**, but also that this constant is zero.

[4] F. Delyon and J. Perière J. Stat. Phys. **64**, 363 (1991); F. Delyon and D. Petritis Comm. Math. Phys. **103**, 441 (1986); F. Delyon and B. Souillard Comm. Math. Phys. **89**, 415 (1983); A. Süto, in *Beyond Quasicrystals*, p.481-549 edited by F. Axel and D. Gratias, (Editions de la Physique, Les Ulis,).

[5] In AlCuFe e.g. the phonons neatly follow the general scheme of broadening, but there is no evidence that would tally with a universal *ansatz* of anharmonicity. A change of slope occurs in the temperature dependence of the Debye-Waller factor, but this is entirely due to the onset of phason hopping dynamics (see e.g. G. Coddens, S. Lyonnard Y. Calvayrac, Phys. Rev. Lett. **78**, 4209 (1997); see also H. de Araújo, A.A. Gomes, and J.B.M. da Cunha, Solid State Comm. **97**, 1025 (1996)).

[6] J. Hafner and M. Krajci, J. Phys.: Cond. Matter **5**, 2489 (1993); M. Krajci and J. Hafner, J. Non-Cryst. Solids, **192-193**, 338 (1995).